RELATIVE GEOLOGIC AGE MEASUREMENTS ON GRANITES BY PLEOCHROIC HALOES AND THE RADIOACTIVITY OF THE MINERALS IN THEIR NUCLEI

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ABSTRACT

In each grain of the radioactive minerals contained in haloes in various Japanese granites, the radioactivity was measured by an autoradiographic method; the development of the pleochroic haloes and the radioactivity of the minerals in their nuclei were both studied in several granites. Their relative geologic ages were determined approximately.

INTRODUCTION

The method of determining the age of minerals from the amount of uranium and thorium contained in them, and the blackness of their pleochroic haloes was devised in 1913 by Joly and Rutherford (1).

The blackness of the halo in biotite increases with the number of alpha-particles emitted from the minute radioactive nuclei; and this in turn depends on the radioactivity and the age of the mineral of the nucleus. As it was impossible, however, at that time to measure the radioactivity of a single mineral grain, Joly's method could not be carried out effectively. The content of the radioactive elements in these minute mineral grains in granite is, in fact, not as uniform as Joly estimated. The result obtained by the present author indicates that grains even in the same thin section of rock are of various species with varying contents of radioactive elements. This will be dealt with in another paper.

By means of the relative blackness of pleochroic haloes, Henderson tried to measure geologic time (2). It was a suggestive method, but apparently not applicable to any rocks younger than 400 million years.

The chief difficulty in these methods of age determination lies in the bleaching or reversal phenomenon. Is this phenomenon really due alone to overexposure to alpha particles, as it has been supposed? Is it impossible to assume that it is rather due to alteration of the biotite itself?

The use of nuclear type plates (3) made the present measurement of radioactivity of minute mineral grains in a thin section much more effective, though the main process of this experiment was but a modification of autoradiography, by which process alone we may measure the radioactivity of a mineral grain so small that it is invisible to the naked eye. The author feels that this method may make some contribution to studies on petrogenesis.

EXPERIMENTAL PROCEDURE

The thin sections used for autoradiography were 30 microns in thickness, as usual, but without cover-glasses. The printed mark method was adopted, as will be stated below, so that the thin section and the developed photo-plate correlate exactly with each other. To spare much of the time needed for the exact correlation of the thin section with the developed plate, the following technique is recommended.

(1) Three corners of the thin section are stained a little with black lacquer.
(2) A cross-shaped window is cut open on each of the three stains (Fig. 1 *a*).
(3) On the other side of the glass is pasted a piece of black paper (Fig. 1 *b*).

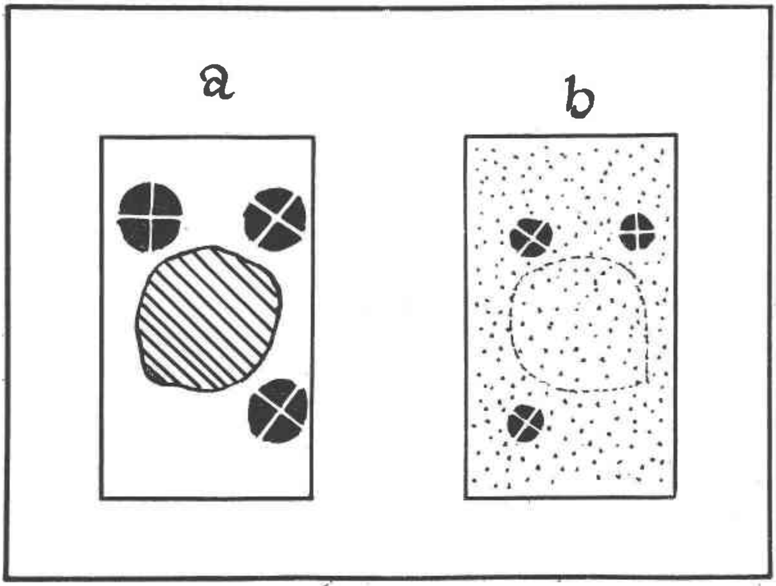


FIG. 1. (*a*). The surface of a thin section of granite with three cross-marks on it. (*b*). The black paper fixed to the reverse of the slide.

(4) Three small holes are cut in the black paper just against the three stains with the cross-shaped windows.
(5) A half-sized nuclear-type plate is fixed to the thin section thus prepared.
(6) The plate is exposed for a while to dim light through the three holes on the black paper. Thus the three cross-shaped windows on the slide are printed on the plate.
(7) The thin section and the plate are kept in contact in a dark place for some four weeks.
(8) The plate is developed with D-19 developer for 12 minutes at ordinary temperatures, and then fixed with F-5.

(9) The black paper is removed from the slide.
(10) A drop of glycerine is placed on the thin section.
(11) The photo-plate is put on this and moved until three cross-shaped marks come into coincidence with the windows on the thin section.
(12) Exact adjustment is possible only under the microscope at 150 ×, for the thickness of the photo-plate hinders the use of any higher magnification. This magnification proved to be most effective for scanning the radioactive centers.

The nuclear type plate used for this purpose was *ET-2E*, 50 microns in thickness, made by the Research Laboratory of the Fuji Photo Film Co., Ltd., of Japan, which was found little inferior in quality to Kodak *NTA*.

The number of the alpha tracks printed on the plate represents the radioactivity of each nucleus mineral grain. No thicker section than 30 microns is needed because of the short ranges of alpha particles within rock minerals.

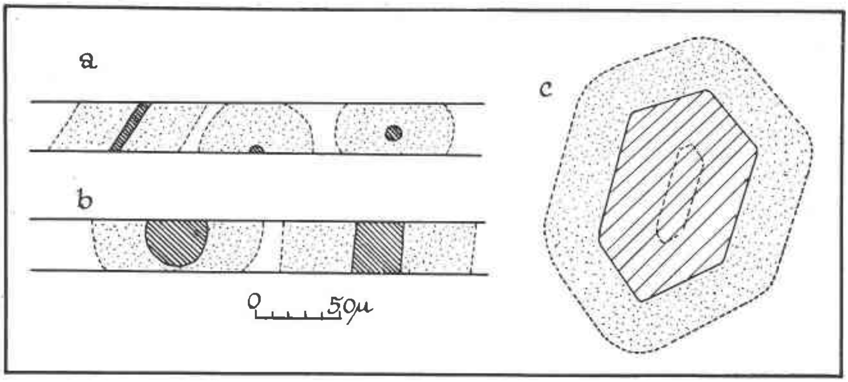
The radius of haloes and the size of the nucleus minerals can be measured by means of a micrometer eyepiece equipped with travelling lines (Schrauben Mikrometer Okular) previously calibrated with a standard micro-scale.

The smaller the size of the nucleus mineral, the more alpha tracks per unit volume are expelled from it, owing to the small absorption of alpha particles. An extremely minute size is no doubt the most desirable, but is inapplicable to the present purpose. The reasons are as follows:

(1) In pleochroic haloes with nucleus minerals smaller than 5 microns in size, the boundary between the halo and its nucleus mineral is hardly distinguishable.
(2) Because of the lack of alpha tracks expelled during a moderate period of exposure, a nucleus mineral too small in size cannot be used.
(3) The nature of the nucleus mineral is very difficult to make out.
(4) As shown in Fig. 2*a*, the number of alpha tracks on the plate does not exactly represent the radioactivity of the nucleus mineral which has blackened the biotite, if their positions and shapes in the thin section differ.

For the determination of the relative geologic ages of two different granites, it is desirable that the greatest possible number of pleochroic haloes available be compared with the radioactivities of their nucleus minerals, which should be, of course, nearly the same in size and shape. The radioactive minerals contained in granites are usually zircon and allanite. Of these two, zircon grains especially are often found to be of moderate and nearly equal size; in radioactivity, however, these minute grains of zircon show great variations, even in the same thin section.

For the comparison, the most desirable nuclei are minute zircons 30



FIGS. 2. (*a*) & (*b*) The transversal section of the thin section. (Note the comparative positions of the pleochroic haloes and their nucleus minerals). (*c*) The plan of the thin section of a radioactive mineral with zonal halo. (The alpha particles, emitted from the central part of the mineral, can not reach the outermost part of the halo).

microns in diameter with disk type pleochroic haloes around them. If they are of this size, not only do their positions in the thin section matter little, but also their alpha particles are not so much absorbed within the zircon grains themselves (Fig. 2*b*).

Crystals larger than 40 microns in diameter and with zonal haloes around them can also be compared with one another (Fig. 2*c*). In this case however, any calibration of the size of the nucleus minerals is unnecessary, as the mineral grain is larger than the longest range of its alpha particles. What is to be compared here between two rocks is (1) the radioactivity in unit area of the polished surfaces of the minerals, and (2) the blackness of the zonal haloes. The results derived from these two methods: disk halo and zonal halo, show a satisfactory agreement.

Varieties of the Minute Minerals in Granitic Rocks

Zircon, allanite, sphene, monazite, xenotime, and the like are the ordinary radioactive accessory constituents of granite. The methods described above enabled the author to discover also in thin sections of granite, uraninite and other highly radioactive minute mineral grains. It has been known for a long time that the nucleus minerals of pleochroic haloes, even though nearly of the same size, differ greatly in the stage of development of their haloes, even in the same flake of biotite. Zircon shows remarkable variations in its radioactivity, and the more radioactive the granite as a whole, the more remarkable is the variation. The author finds that the more radioactive the granite, the more haloes it offers for comparing.

THE RELATION OF THE RADIOACTIVITY OF THE NUCLEUS MINERAL TO THE STAGE OF ITS HALOES

The blackening of biotite by alpha particles represents a complicated process, but the application of the autoradiographic technique sheds considerable light on this subject. Figure 3 illustrates the results on granite from Tanakamiyama, Siga Prefecture, Japan; the ordinate is the radii of its haloes in microns, the abscissa is the alpha particles ejected per day from one grain of the nucleus mineral.

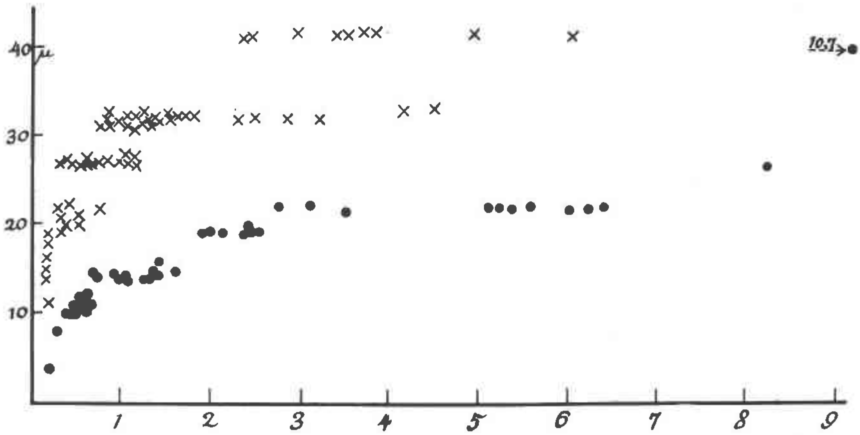


FIG. 3. The relation of halo radii and the radioactivities: Tanakamiyama granite (crosses) and Masutomi granite (solid circles). The size of nucleus mineral: about 30 microns in diameter. Ordinate: halo diameter in microns. Abscissa: alpha radioactivity from one grain of mineral per day. (All crosses from Fig. 3 to Fig. 7 represent Tanakamiyama granite).

The nucleus minerals, mostly zircon, were 25–35 microns in diameter; halo rings were measured with a micrometer ocular, using their outermost radii. The marks shown on the levels of 41, 32, and 27 microns represent the ThC′, RaC′ and ThA rings respectively; while the rings smaller than 23 microns represent complicated overlaps of the various rings. Though the density of the haloes is not given in the figure, be it noted that in the case of haloes of similar radius, the blackness is deeper on the right-hand side than on the left. The left sides of haloes are generally either embryonic or under-exposed, the right sides are over-exposed. In the semiquantitative measurement of the relative ages of two rocks, however, the best method to use is to compare the two radioactivities at the stage of the first slight appearance of their haloes. Is it possible here that, as the biotites differ, the blackness of the haloes in question may also differ? So far as the granite of Tanakamiyama, containing two sorts of biotites, is concerned, a difference of this kind appears to be negligible. One biotite is brown, with refractive index $(\gamma)=1.64$, and is centrally located; for the other, greenish in color and myrmekitic in structure, zonally surrounding the first, and presumably containing much fluorine, $(\gamma)=1.63$. The haloes in these two biotites show no difference in their degree of blackness. Even in the case of two different granitic rocks, a similar result also may be expected. The experimental projection of numerous alpha particles on various biotites will perhaps verify this view.

According to some investigators, the bleaching or reversal phenomenon in biotite, which is inevitable for pleochroic haloes, is extremely irregular and complicated. The radiographic studies of the author showed no such irregularity, especially in the case of granites whose age was younger than Mesozoic. Besides, it is a noteworthy fact that the reversed halo of what G. H. Henderson called "extinct halo type A" (10) is seen in the xenoliths of the Tanakamiyama granite (4). This extinct halo, however, seems to the author to be due rather to the alteration of the biotite itself than to over-exposure by alpha particles from nucleus minerals, as has been believed. Though the author's results are limited in number, it seems probable that what is true of extinct haloes will some day be found true of haloes in general.

On Pleochroic Haloes in Various Japanese Granites

In South Siga there is a stock, called the Tanakamiyama granite, which outcrops in an ellipse about 14 km. long by 7 km. wide. This unconformably contributes pebbles to the Kobiwako Plio-Pleistocene formation. The granite is mostly coarse-grained, with biotite, but in its marginal phases is fine- or medium-grained, or sometimes passes into porphyritic rocks. It is generally rich in radioactive elements. According to T. Asayama's measurements (5) the radium content varies from 3.23×10^{-12} g/g to 1.48×10^{-12} g/g. This radium content seems rather high for Japanese granitic rocks according to the data of Z. Hatuda (6, 7).The biotite in this district contains many pleochroic haloes and especially ThC′ haloes. On account of the slight diversity in shape and size of the minute mineral grains, some deviation from an exact linear relation is almost inevitable, for only in the case of grains of exactly the same size and shape, can exact comparison of blackening of the haloes be established. The haloes in the altered biotite are not suitable for a standard of comparison.

In Fig. 3 the dots indicate the granite in the Masutomi Spring district, Yamanashi Prefecture. Its intrusion into the Misaka formation (Miocene sediments) tells its age. The springs of this district are noted for their high radioactivity (8, 9) and the granites nearby are also fairly radioactive. With respect to the order of radioactivity of the minute crystals of accessory minerals the Masutomi and Tanakamiyama granites show no marked difference, but as far as the stages of the haloes are concerned, the former appears younger than the latter. The pleochroic haloes are, in Henderson's (10) terminology, mostly active ones; since the formation of the minerals, alpha particle ejection must have remained constant. When more than one million years have passed, the mineral has reached radioactive equilibrium.

Now let the ages of two granites A and B be Y_A and Y_B years, and the radioactivities of their nucleus minerals (similar in shape and size) be

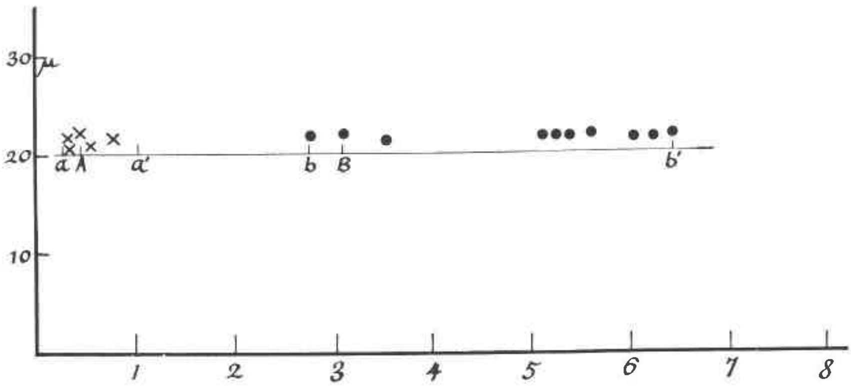


FIG. 4. Haloes about 22 microns in diameter. (See Fig. 3).

R_A and R_B (Fig. 3). If the pleochroic haloes originating from these two nucleus minerals show the same stage of blackening, then $Y_A/Y_B = R_B/R_A$.

The ratio of the two geologic ages, therefore, is inversely proportional to the radioactivity. In other words, if the stages of development of a halo and the radioactivity of its nucleus mineral can be measured, the relative geologic ages of the rocks in question can be determined. Henderson's apparatus (11), for instance, would enable us to measure the halo blackening with greater precision than is possible in the present instance. As an example, pleochroic haloes about 22 microns in radius may be considered. In Fig. 4, the aAa' group represents the Tanakamiyama granite, the bBb' the Masutomi, and the points a and b represent the faintest haloes barely recognizable under the microscope, while a' and b' are the most deeply blackened ones. The following relations are formulated, for simplicity's sake, without considering the composite haloes of the Th and U series combined:

$$a/b = A/B = a'/b' = (a' - a)/(b' - b) = Y_B/Y_A.$$

Any two corresponding points on the line aa' and bb' representing the relative blackness of haloes give the required ratio. The age is represented by $a' - a/b' - b$, hence the younger the haloes are, the greater is the difference in their radioactivities.

This tendency is not necessarily limited to certain specific haloes. RaC′ and ThC′ haloes are preferable for comparison, as they form the outermost rings of the compound haloes of the U and Th series, respectively. If a stricter comparison is necessary, the color, thickness, and orientation of the biotite in the thin section must also be considered. Halo A (Tanakamiyama granite) and halo B (Masutomi granite) are nearly the same in blackness. Hence, roughly speaking:

$$Y_A/Y_B = B/A = 3/0.5 = 6/1.$$

On this basis, because halo B belongs, as stated above, to the Miocene; using the latest geologic time scale (12), halo A should be nearly 6 times as old, and be ascribed to the middle Cretaceous. No other geological datum for the Tanakamiyama granite is in discord with this result. Figure 5 shows the San-in district granite. The data are still too scanty to tell with exactitude anything more than that its age lies between middle Cretaceous and Miocene, presumably in the oldest Tertiary.

In Fig. 6, the triangles representing the Bofu granite of Yamaguchi Prefecture are further to the right of the diagram than those representing

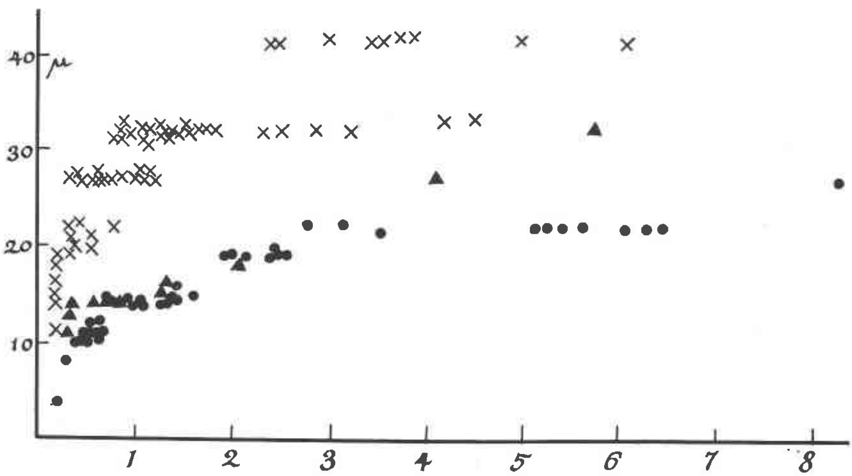


FIG. 5. San-in district granite (triangles) represents an age between Miocene and middle Cretaceous.

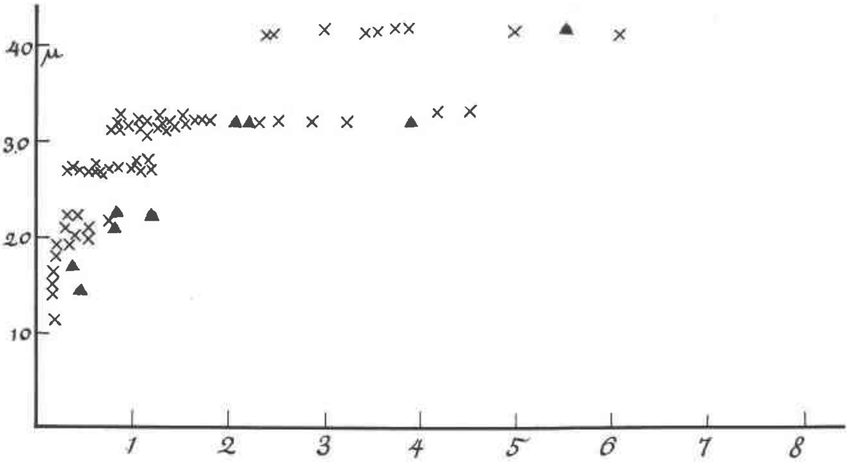


FIG. 6. Triangles: Bofu granite (Yamaguchi prefecture).

the Tanakamiyama granite, and this tells us that the intrusion of the Bofu took place a little later (upper Cretaceous?) than that of the latter. Similar results were obtained for granites from Kitashirakawa, Kyoto Prefecture; Ookawara, Shiga Prefecture; Hoki, Kyoto Prefecture; and Katanoyama, Osaka Prefecture.

The zonal halo method is applicable only to a mineral in which the radioactive elements are equally and homogeneously distributed. Such haloes are neither rare nor exceptional. This method is superior to the other one, for it requires no calibration of the size of a nucleus mineral. The results obtained by this method are shown in Fig. 7. The radioactivity is shown on the abscissa as the number of alpha particles emitted from the polished surface per day per 0.01 $mm.^2$ by minerals such as coarse-grained allanite and zircon. In either method, however, great care is needed in the following:

(1) The size of the halo must be measured under the microscope as exactly as possible.
(2) No altered biotite containing pleochroic haloes can be used.
(3) In halo measurements, the position and the orientation of the radioactive mineral grains as shown in the thin section must be taken into consideration.

So far as a specimen of biotite containing a pleochroic halo is *fresh*, any two rocks may be compared to determine their relative geologic ages, whether the rock be pegmatite, volcanic rock, mica schist, or any other.

Under favorable circumstances, the order of age of two rocks can easily

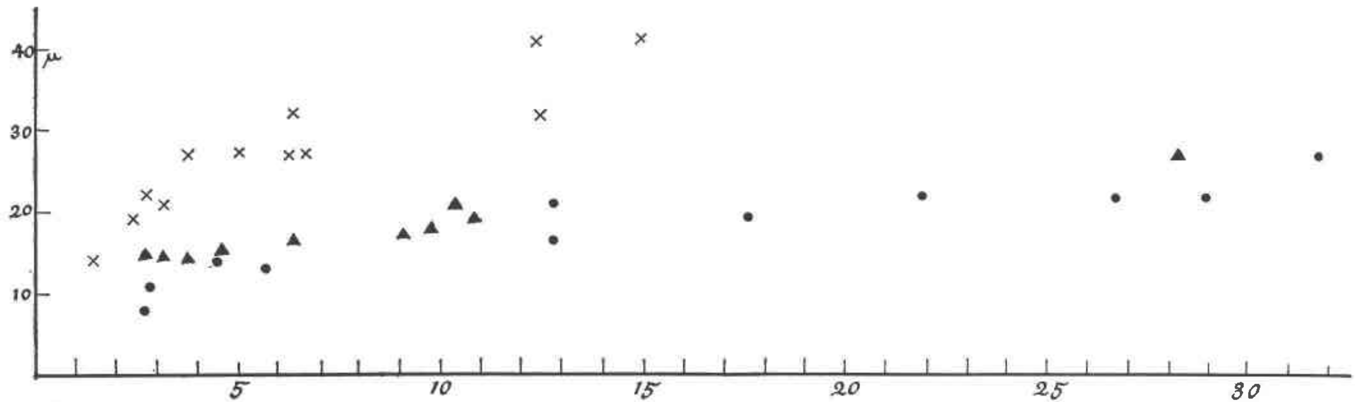


FIG. 7. The relation between width of a zonal pleochroic halo and its radioactivity. Tanakamiyama granite (crosses), San-in granite (triangles), and Masutomi granite (solid circles). Ordinate: halo width in microns. Abscissa: alpha radioactivity from the polished surface per 0.01 mm.^{2} per day.

be determined even if they were of successive intrusions. Suppose that the same type of alpha particles gave various degrees of blackness to different biotites. If such be the case, the method of blackening the biotite artificially by exposure to a given source of alpha particles of known strength would enable us to determine not only the relative ages of two rocks, but even give us some approximation to their absolute ages. And, when autoradiographic studies become more advanced, even one pleochroic halo would, under favorable conditions, be enough to determine the age of a given rock.

Among the advantages of the author's method of measuring ages are the facts that it is freer from contamination difficulties than the lead method, and from difficulties of alteration than the helium method. Pleochroic haloes may some day be a more exact index of the age of igneous or metamorphic rocks than fossils are in sedimentary rocks.

Summary

(1) Even in the same specimen of granite the minute radioactive mineral grains vary in activity.

(2) The blackness of a pleochroic halo is proportional to the product of the radioactivity and the age of the nucleus mineral.

(3) The relative geologic age of two rocks each bearing pleochroic haloes is easily determined by comparing the stages of development of the haloes in each, and by contrasting the respective radioactivities of the nucleus minerals of the haloes.

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